

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re Application of:

Marina E. Kondakova, et al

ORGANIC  
ELECTROLUMINESCENT DEVICES  
WITH ADDITIVE

Serial No. US 10/729,737

Filed 05 December 2003

Group Art Unit: 1774

Examiner: Dawn L. Garrett

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA. 22313-1450

Sir:

**THIRD DECLARATION OF MARINA E. KONDAKOVA  
UNDER 37 CFR 1.132**

The undersigned, Marina E. Kondakova, declares that:

She has received the degree of M.Sc., Colloid and Surface Chemistry from St. Petersburg University, St. Petersburg, Russia in 1987 and Ph.D., Physical Chemistry from St. Petersburg Technological University of Plant Polymers, St. Petersburg, Russia in 1993;

Since 1993 she has been employed as a research scientist with the Institute for Molecular Science, Okazaki Japan, then with the Research Institute of the Pulp and Paper Industry, St. Petersburg, Russia, and, since 2002, with the Display and Components OLED Materials R&D Group of Eastman Kodak Company;

She is an inventor in the above-captioned patent application;

She has reviewed the outstanding Office Action and any applicable cited references;

Under her direction and control, samples were prepared and experiments conducted and the results were recorded as follows:

#### Experimental Examples 8-13:

Device 8, not satisfying the requirements of the invention, was fabricated in an identical manner to comparative Device 5 at page 45 of the specification, with the following exceptions. The thickness of the indium-tin oxide (ITO) layer was 25 nm, rather than 85 nm. The material of the hole-blocking layer (HBL) was BA1q1 rather than BA1q. This BA1q1 is bis(2-methylquinolin-8-olato) (2,6-diphenyl-phenolato) aluminum(III), a compound quite similar to BA1q. Instead of an Mg:Ag cathode, a bilayer made of 0.5 nm of lithium fluoride followed by 100 nm of aluminum was vacuum deposited onto the ETL. Thus, Device 8 has the following layered structure: ITO | CF<sub>x</sub> (1 nm) | NBP (75 nm) | CBP + 6 wt. % Ir(ppy)<sub>3</sub> (35 nm) | BA1q1 (10 nm) | Alq (40 nm) | LiF | Al.

Devices 9-11, satisfying the requirements of the invention, were fabricated in an identical manner to Device 8 except that the concentration of MTDATA in the LEL was as indicated below in Table 4. Devices 12 and 13, for their particular concentrations, do not provide an improvement over Device 8.

The devices thus fabricated were tested for efficiency and color at an operating current density of 6 mA/cm<sup>2</sup>. The results, including 1931 CIE (Commission Internationale de l'Éclairage) coordinates, CIE<sub>x</sub> and CIE<sub>y</sub>, are reported in Table 4.

Table 4. Evaluation Results for Devices 8 through 13.

Device	MTDATA (wt. %)	Luminous yield (Cd/A)	Power efficiency (lm/W)	CIE <sub>x</sub>	CIE <sub>y</sub>	Type
8	0	31.9	12.3	0.291	0.626	Comparison
9	3	24.5	7.4	0.293	0.619	Invention
10	10	38.1	14.5	0.289	0.625	Invention
11	15	37.1	14.5	0.287	0.626	Invention
12	20	23.6	10.4	0.284	0.619	Comparison
13	30	7.41	3.7	0.279	0.590	Comparison

As can be seen from Table 4, inventive Devices 10 and 11, containing the efficiency-enhancing material MTDATA at 10 and 15 wt. %, respectively, exhibit superior performance relative to comparative Devices 8, 12, and 13 in which the efficiency-enhancing material is either absent or is present in amounts that are not effective compared to Device 8.

#### Experimental Examples 14-17:

An EL device (Device 14) satisfying the requirements of the invention was constructed in the following manner:

1. A glass substrate, coated with an approximately 25 nm layer of indium-tin oxide (ITO) as the anode, was sequentially ultrasonicated in a commercial detergent, rinsed in deionized water, and exposed to an oxygen plasma for about 1 minute.
2. Over the ITO was deposited a 1 nm fluorocarbon ( $\text{CF}_x$ ) hole injecting layer (HIL) by plasma-assisted deposition of  $\text{CHF}_3$  as described in US 6,208,075.
3. Next, a hole transporting layer (HTL) of *N,N'*-di-1-naphthyl-*N,N'*-diphenyl-4,4'-diaminobiphenyl (NPB) was vacuum deposited to a thickness of 50 nm.
4. A 50 nm light emitting layer (LEL) consisting of a mixture of 4,4'-*N,N'*-dicarbazole-biphenyl (CBP) as a host, *fac*-tris(2-phenylpyridinato) iridium (III) [i.e.,  $\text{Ir}(\text{ppy})_3$ ] at 6 wt. % as a green phosphorescent emitter, and 4,4',4''-tris(*N*-3-methylphenyl-*N*-phenyl-amino)triphenylamine (MTDATA) at 10 wt. % as an efficiency-enhancing material was then vacuum deposited onto the hole-transporting layer.
5. A hole-blocking layer (HBL) of bis(2-methylquinolin-8-olato) (2,6-diphenyl-phenolato) aluminum(III) ( $\text{BAIq1}$ ) having a thickness of 10 nm was vacuum deposited over the LEL.
6. An electron transport layer (ETL) of tris(8-quinolinolato) aluminum (III) ( $\text{Alq}$ ) having a thickness of 60 nm was vacuum deposited over the HBL.
7. 0.5 nm of lithium fluoride was vacuum deposited onto the ETL, followed by a 100 nm layer of aluminum, to form a bilayer cathode.

The above sequence completed the deposition of the EL device. Therefore, Device 14 had the following layer structure: ITO |  $\text{CF}_x$  (1 nm) | NPB (50 nm) | CBP + 6 wt. %  $\text{Ir}(\text{ppy})_3$  + 10 wt.% MTDATA (50 nm) |  $\text{BAIq1}$  (10 nm) |  $\text{Alq}$  (60 nm) | LiF | Al. The device, together with a desiccant, was then hermetically packaged in a dry glove box for protection against ambient environment.

A comparative Device 15 not satisfying the requirements of the invention was fabricated in an identical manner to Device 14 except that MTDATA was not included in the LEL.

A comparative Device 16 not satisfying the requirements of the invention was fabricated in an identical manner to Device 14 except that the HTL of NPB was omitted. Device 16 had the following layer structure: ITO | CF<sub>x</sub> (1 nm) | CBP + 6 wt. % Ir(ppy)<sub>3</sub> + 10 wt.% MTDATA (50 nm) | BAq1 (10 nm) | Alq (60 nm) | LiF | Al.

A comparative Device 17 not satisfying the requirements of the invention was fabricated in an identical manner to Device 14 except that the HTL was omitted and MTDATA was not included in the LEL. Device 17 had the following layer structure: ITO | CF<sub>x</sub> (1 nm) | CBP + 6 wt. % Ir(ppy)<sub>3</sub> (50 nm) | BAq1 (10 nm) | Alq (60 nm) | LiF | Al.

The devices thus fabricated were tested for efficiency and color at an operating current density of 6 mA/cm<sup>2</sup>. The results, including 1931 CIE (Commission Internationale de l'Éclairage) coordinates, CIE<sub>x</sub> and CIE<sub>y</sub>, are reported in Table 5.

Table 5. Evaluation Results for Devices 14 through 17.

Device	HTL	MTDATA (wt.%)	Luminous yield (Cd/A)	Power efficiency (lm/W)	CIE <sub>x</sub>	CIE <sub>y</sub>	Type
14	NPB	10	23.5	6.0	0.339	0.604	invention
15	NPB	0	19.9	5.3	0.338	0.605	comparison
16	none	10	17.2	5.0	0.351	0.597	comparison
17	none	0	17.3	4.8	0.352	0.597	comparison

As can be seen from Table 5, inventive Device 14 including both a HTL (NPB) and an efficiency-enhancing material in the LEL (10% of MTDATA) demonstrates an improved luminous yield and power efficiency relative to comparative Device 15 that lacks an efficiency enhancing material. Inventive Device 14 is also superior to Comparative Device 16, which lacks a HTL, and to Comparative device 17, which lacks both a HTL and an efficiency-enhancing material.

Examples 14-17 were constructed to demonstrate the necessity of using both a HTL and an efficiency-enhancing material in the LEL in order to achieve a substantial improvement of luminous yield and power efficiency. The thicknesses of the organic layers in inventive Device 14 were not optimized to obtain the best possible values. Therefore, the performance of the inventive Device 14 differs from performance of

the optimized inventive Device 10 shown in Table 4. I did not expect such an improvement from the combined presence of the HTL and the efficiency enhancing material.

All statements made herein of my knowledge are true, and all statements made on information and belief are believed to be true. These statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this 14<sup>th</sup> day of September, 2007

M. Kondakova

Marina E. Kondakova